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ORGANOMETALLIC LIQUIDS AS POTENTIAL DOSIMETRIC MATERIALS: A PULSE RADIOLYSIS STUDY OF THE TETRA-ALKYL COMPOUNDS OF Si, Sn AND Pb

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June 1976

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The methods of kinetic optical spectroscopy have been employed to observe the very short-lived transients produced by pulse radiolysis of some of the tetra-alkyl compounds of Si, Sn and Pb doped with naphthalene and toluene. The feasibility of utilizing such bimolecular compounds for time resolved dosimeters in the low-energy x-ray region has been demonstrated. This represents an additional method for

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20. ABSTRACT (continued)

refinement of the pulsed radiation profiles such as might be encountered in nuclear weapons uses.

SUMMARY
(Nontechnical)

A continued requirement exists to develop dosimetry systems suitable for use in pulsed fields of ionizing radiation, particularly in the low-energy x-ray region. The desirable characteristics of such dosimeters should include both a sensitivity to the spectral distribution of the incident radiation and a capability of live time readout to follow pulse profiles over a time scale of several nanoseconds.

The organometallic compounds of the Group IVA metals have been shown to produce optically detectable changes after exposure to ionizing radiation. Theoretically, the K-shell energy edges of their metal atoms can serve as windows to determine the spectral distribution of the radiation pulse. The long lifetimes of the radiation products, however, preclude their use to determine the time domain profile of the radiation pulse.

This phase of the investigation was undertaken to assess the feasibility of doping the pure organometallic liquids with aromatic compounds, such as naphthalene and toluene, to produce short-lived radiation-induced effects without significantly altering the energy dependent properties of the organometallic matrix. Very intense photon emission on time scales comparable to the radiation pulse was observed under conditions which do not affect the trialkyl-metal bond scission necessary for energy discrimination.

TABLE OF CONTENTS

	Page
Summary (Nontechnical)	1
Introduction	5
Methods and Materials	5
Results and Discussion	6
Conclusions	8
References	10

LIST OF FIGURES

Figure 1.	Oscilloscope trace of naphthalene-doped Et_4Si	6
Figure 2.	Long-lived optical absorption transients at 320 nm of pure Et_4Si	7
Figure 3.	Long-lived optical absorption transients at 320 nm of naphthalene-doped Et_4Si	8

INTRODUCTION

In an earlier publication,[2] it was observed that the tetra-alkyl compounds of the Group IV elements silicon, germanium, tin, and lead exhibited transient absorption spectra in the visible and near ultraviolet regions following radiolysis. The lifetimes of these spectra varied from a few to hundreds of microseconds. It was suggested that if it were possible to reduce the lifetimes of the transient species by the addition of a suitable reactant to the target material it might be possible to monitor the profile of a radiation pulse with a rise and fall time of several nanoseconds.

Toluene and naphthalene were selected as reactants to test this hypothesis.

METHODS AND MATERIALS

The experimental procedure was essentially that described in detail previously.[2] Briefly, the AFRRI electron linear accelerator was used as a pulsed source of ionizing radiation. The organometallic liquids were exposed to single pulses of 40 MeV electrons, each of 200-nsec duration, selected from a 10 percent energy bin. Changes in optical absorption of the irradiated material were examined by the methods of kinetic spectroscopy. Variations in the intensity of the analyzing light source, detected as a photodetector voltage, were monitored on an oscilloscope and photographed. Minor electronic changes of the electronic detection system described previously reduced the apparatus time constant to $\sim$ 100-200 nsec.

Tetraethyl silane (Et_4Si), purchased from PCR, Inc., tetraethyl tin (Et_4Sn), purchased from K & K Laboratories, Inc., and tetramethyl lead (Me_4Pb) purchased from Ventron Corporation, Alfa Products, were used for this study. The tetraethyl silane and tin were doped with 7.7×10^{-5} moles/liter of naphthalene, Fisher certified. The tetramethyl lead was purchased as 80 percent in toluene, which was not removed. After doping, each solution was deaerated by bubbling with nitrogen, ultrapure grade, to remove dissolved oxygen. Spectroscopic grade toluene was purchased from Fisher Scientific Company.

A Faraday cup, placed in line of sight behind the irradiation cell, was used to monitor the LINAC beam current. The aqueous ferrocyanide dosimeter was used to

determine the dose deposited in H_2O in the same irradiation cell used for the organometallic liquids. The LINAC beam parameters were chosen to be those which would deposit 1030 ± 30 rads/pulse into the ferrocyanide solution.

RESULTS AND DISCUSSION

Each of the three organometallic compounds displayed very intense, short-lived, light emission in visible and near ultraviolet regions following radiolysis. The photodetection system was saturated at all voltages in the linear response region by the light emission. A typical oscilloscope trace of Et_4Si, doped with 7.7×10^{-5} moles/liter naphthalene, is shown in Figure 1. The wavelength of detection is 320 nm. The

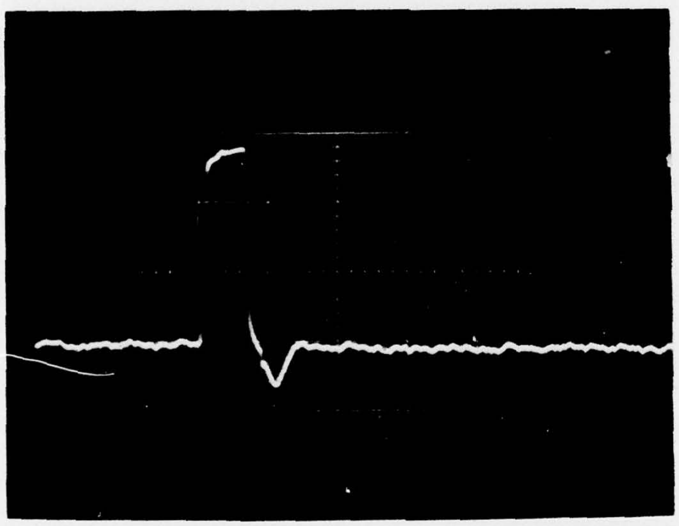

Figure 1. Oscilloscope trace of naphthalene-doped Et_4Si. Vertical scale 1 V/cm; horizontal scale 1 μsec/cm.

vertical scale of this figure is 1 V/cm and the horizontal scale is 1 μsec/cm. The increase in voltage corresponds to detection of a light pulse at least twice as intense as the brightness of the xenon lamp analyzing light source. The relatively slow recovery of the overloaded optical and electronic analyzing system does not permit kinetic analysis of the decay of the light emission. It should be noted, however, that the decay is time-constant limited.

The 0.64-volt "overshoot" signal, peaking at 1.2 μsec after the start of the intense light pulse, may be a result of the incomplete recovery of the electronic detection system. If it were due to light absorption by a radiation-induced transient product, the signal would correspond to an optical density charge of 0.139 (4 cm), some 18 times as intense as the long-lived absorption observed at this wavelength in pure Et_4Si. Neither the very intense light pulse nor the overshoot is observed in irradiated pure Et_4Si. Similar light emission and overshoot signals were observed in naphthalene-doped Et_4Sn and Me_4Pb containing toluene. Neither pure Et_4Sn nor pure toluene displayed these light emissions nor intense absorptions after exposure to pulses of ionizing radiation.

The population of excited states of aromatic compounds has been used to study radiation-induced changes in low temperature organic glasses.[1] The formation of the excited states and their decay by photon emission can be used in the liquid state to produce very short-lived detectors of radiation damage to the host matrix. The lifetime and intensity of the light emission are dependent on the recombination rate and, hence, the concentration of the fluor.

The long-lived optical absorption transients at 320 nm are shown in Figures 2 and 3. Figure 2 is the oscilloscope trace of pure Et_4Si and Figure 3 is that of

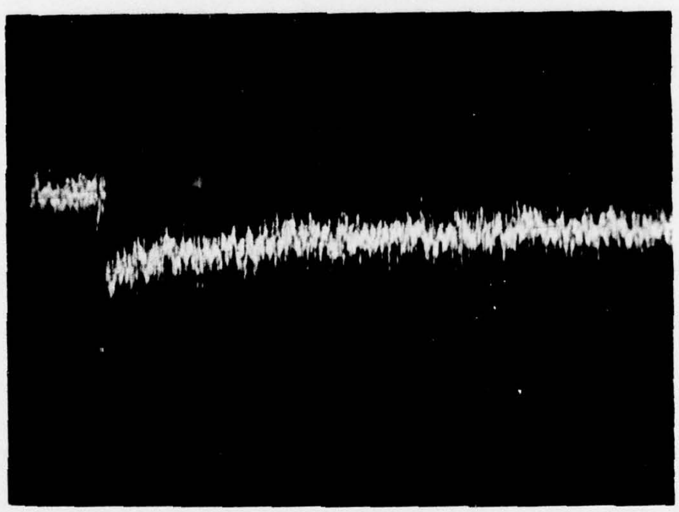

Figure 2. Long-lived optical absorption transients at 320 nm of pure Et_4Si. Vertical scale 50 mV/cm; horizontal scale 50 μsec/cm.

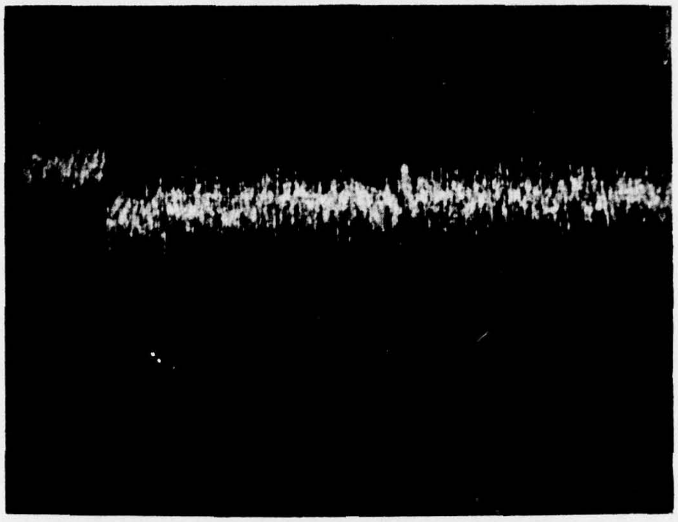

Figure 3. Long-lived optical absorption transients at 320 nm of naphthalene-doped Et_4Si. Vertical scale 50 mV/cm; horizontal scale 100 μsec/cm.

naphthalene-doped Et_4Si. The vertical scale in each case is 50 mV/cm and the horizontal scales are 50 and 100 μsec/cm, respectively. The initial optical density of the radiation-induced transient is 0.0073 (4 cm) for the pure Et_4Si and 0.0075 (4 cm) for the naphthalene-doped Et_4Si, indicating that the ionizing radiation is primarily absorbed by the organometallic matrix. The kinetics of the decay pattern, which seem to differ during the first 20-30 μsec due to the different oscilloscope sweep speeds and electronic integrating constants, are quite similar. Had the aromatic compound, in this case naphthalene, served as an efficient trap for the ionizing radiation, the concentration and the kinetics of decay of the much longer lived trialkyl-metal free radicals[2] would have been significantly altered.

CONCLUSIONS

The use of the organometallic compounds as dosimeters in the low-energy x-ray region is based on their energy absorbing properties, i.e., the ratio of photoelectric to Compton energy absorption is large. The primary obstacle to time domain resolution of a radiation pulse is due to the relatively long lifetimes of the trialkyl-metal

radicals. The addition of small quantities of aromatic compounds to these organometallic liquids does not appreciably alter the radiolysis product concentration but does produce an excited state, in naphthalene, which decays on a time scale comparable (within a factor of two) to the duration of the radiation pulse.

The sensitivity of the matrix to the energy spectrum of the incident radiation is unaltered but the ease of detection is significantly increased. Although a detailed investigation to determine the optimum parameters lies beyond the capabilities of the presently used detection system, the feasibility of using such bimolecular solutions to characterize the profile of a radiation pulse has been demonstrated.

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